The Nernst-Planck Equation in Cylindrical Space and Time Coordinates for the Chloride-Ion Transport Phenomenon in Concrete Materials Using Theoretical Study

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Abstract

The effective evaluation of transport phenomenon of chloride-ions in concrete is required in any study of the durability problem of concrete members and structures. In this paper, the analytical solution of the Nernst-Planck equation in cylindrical space with radial symmetry and time coordinates compared with the results of the accelerated chloride migration test (ACMT) obtained from the literature were used to investigate the transport phenomenon of chloride-ions in concrete. The diffusion/migration coefficient of chloride ion was calculated by the analytical solution of the proposed method and compared with the previous results of the ACMT. The results of the present study show that the pozzolanic materials such as fly ash and slag could reduce the chloride permeability in concrete. Both the chloride concentration and migration rate versus time obtained by the calculation of the Nernst-Planck equation in two-dimensional cylindrical space and time coordinates are distinguishably in good agreement with the results measured from the experiment of the ACMT taken from the literature.

***Key Words*:** Acceleration Chloride Migration Test, Mineral Admixtures, Nernst-Planck Equation, Transport Phenomenon

1. Introduction

Durability problem is an important topic for concrete or reinforced concrete (RC) structures exposed to aggressive environments [1,2]. Many environmental phenomena are significantly influencing the durability of RC structures [3–5]. Chloride-ion ingress is one of the major factors to cause structure deterioration. The biggest cause of reinforcement corrosion stems from the depassivation of steel by chloride-ions [6–8]. To treat the deterioration problem of concrete structures due to corrosion, the reducing of permeability of detrimental materials such as Cl^- and SO_4^{2-} in concrete is a good strategy. When the migration rate of harmful ions in concrete is reduced the durability of concrete is increased. Therefore, the effective evaluation of transport behavior of chloride-ions in concrete material is a very important research topic.

The conventional chloride diffusion testing method needs considerable time to obtain the steady-state chlo-

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ride flow across the specimen. Because conventional chloride diffusion test for the concrete is time-consuming, the rapid chloride penetration test (RCPT) (ASTMC 1202-97 [9] or AASHTO T277-96 [10]) used a potential of 60 V power source and the solution volume 250 ml of each acrylic cell, the current passing through the concrete specimen in 6 hr is measured, and the resistance of the concrete specimen to chloride-ions penetration is evaluated by the total charge passed [11]. Andrade [12], Cho [13], Cho et al. [14,15], Yang et al. [16,17], Yang [18], Yang and Cho [19,20], and Ou [21] used the Nernst-Planck equation in unidirectional space and time coordinates to study the transport phenomenon of chloride-ion in concrete specimen by means of accelerated chloride migration test (ACMT).

Although these studies have provided much valuable information on the chloride diffusivity in concrete with or without mineral admixture using the RCPT or ACMT method, there are still many factors that have not yet been explored. The objective of this paper is to derive the analytical solution of the Nernst-Planck equation in cylindrical space and time coordinates to study the transport phenomenon of chloride-ion in concrete with mineral admixture. The needed parameters are cited from the available test data [13] and taken from the available literature [14–16]. The results of this study may offer a method to provide indication of durability for concrete or RC structures.

2. Theoretical Model

For the nonsteady-state case of diffusion and migration, the Nernst-Planck equation with initial and boundary conditions in two-dimensional cylindrical space with radial symmetry (i.e., suppose there is axisymmery so that chloride concentration does not very with θ and the thus three-dimensional cylindrical space reduces to two-dimensional one.) and time coordinates can be expressed as

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r}\frac{\partial C}{\partial r} + \frac{\partial^2 C}{\partial z^2}\right) - Da\left(\frac{\partial C}{\partial r} + \frac{\partial C}{\partial z}\right) \tag{1-a}$$

$$C(r, z, 0) = 0 \tag{1-b}$$

$$C(r, 0, t) = C_0 \tag{1-c}$$

$$C(r, z \to \infty, t_m) = 0 \tag{1-d}$$

where $a = \frac{z_i FE}{RT}(1/cm)$, z_i is the ion valance, F is the Faraday constant (96500 coulombs/mole), $E = \frac{\Delta V}{l}$ is the electrical field (Volt/cm), l is the specimen length (cm), ΔV is the potential drop across the specimen, R is the gas constant (8.31 Joule/(mole · K)), T is the temperature in Kelvin (K), C(r, z, t) is the pore solution concentration (mole/cm^3) at the position of radius r and z-axis and time t, C_0 is the pore solution concentration (mole/cm^3) at the surface, and D is the diffusion and migration coefficient (cm^2/sec).

Using the method of separation of variables [22], the chloride-ion concentration can be expressed as

$$C(r, z, t) = R(r)Z(z)T(t) \tag{2}$$

where R(r), Z(z) and T(t) represent the physical meanings of radical, thick, and time effects, respectively. Inserting Eq. (2) into Eq. (1-a) and then treating, we get

$$\frac{DR'' + (r^{-1} - Da)R'}{R} + \frac{DZ'' - DaZ'}{Z} = \frac{\dot{T}}{T} = -K_1 \tag{3}$$

where K_1 is the separation constant and is of physical unit in 1/sec.
From Eq. (3),we now have

$$\dot{T} + K_1 T = 0 \tag{4}$$

and

$$\frac{DR'' + (r^{-1} - Da)R'}{R} = -K_1 - \frac{DZ'' - DaZ'}{Z} = K_2 \tag{5}$$

where K_2 is the separation constant and is of physical unit in 1/sec.
Hence, we have

$$DR'' + (r^{-1} - Da)R' - K_2 R = 0 \tag{6}$$

and

$$DZ'' - DaZ' + (K_1 + K_2)Z = 0 \tag{7}$$

Now considering the initial and boundary conditions, we obtain

$$C(r,Z,0) = R(r)\,Z(z)\,T(0) = 0 \Rightarrow Z(z) = 0, T(0) \neq 0, R(r) \neq 0 \quad \text{(8-a)}$$

$$C(r,0,t) = R(r)\,Z(0)\,T(t) = C_0 \Rightarrow Z(z) = C_0, R(r) \neq 0, T(t) \neq 0 \quad \text{(8-b)}$$

and

$$C(r,z,t_M) = R(r)\,Z(z)\,T(t_M) = 0 \Rightarrow Z(z) = 0, R(r) \neq 0, T(t_M) \neq 0 \quad \text{(8-c)}$$

Eq. (4) can be written as

$$\frac{\dot{T}}{T} = -K_1 \quad (9)$$

After integrating to Eq. (9), we have

$$T(t) = \alpha e^{-K_1 t}, \ \ \alpha = \text{constant}, \ \ \alpha = T(0) \neq 0 \quad (10)$$

Eq. (10) can be changed as

$$T(t) = T(0) e^{-K_1 t} \quad (11)$$

Now consider Eq. (7) and its boundary conditions according to Eqs. (8-b) and (8-c)

$$DZ'' - DaZ' + (K_1 + K_2)Z = 0, \ \ Z(0) = C_0, \ \ Z(z) = 0 \quad (12)$$

From Eq. (12), we have

$$Z'' - aZ' + \frac{K_1 + K_2}{D} Z = 0 \quad (13)$$

Eq. (13) is an ordinary differential equation. Let us seek a solution in the form $Z(z) = e^{mz}$, where m is a constant that may be complex. Thus, the characteristic equation of Eq. (13) is

$$m^2 - am + \frac{K_1 + K_2}{D} = 0 \quad (14)$$

The solution of Eq. (14) is

$$m = \frac{a \pm \sqrt{a^2 - 4\dfrac{(K_1 + K_2)}{D}}}{2}, \ (\text{m} = \text{m}_1(+), \text{ and } \text{m}_2(-)) \quad (15)$$

where m_1 and m_2 are constants and of physical units in 1/m.

The general solution of Eq. (13) is

$$Z(z) = P_1\ e^{m_1 z} + P_2\ e^{m_2 z} \quad (16)$$

where P_1 and P_2 are arbitrary constants.

Now consider boundary conditions

$$Z(0) = P_1 + P_2 = C_0 \Rightarrow P_2 = C_0 - P_1 \quad (17)$$

$$Z(z \to \infty) = P_1\ e^{m_1 z} + P_2\ e^{m_2 z} = 0 \quad (18)$$

Thus, we obtain

$P_1 = 0$ and $P_2 = C_0$

The particular solution of Eq. (12) is

$$Z(z) = C_0\ e^{m_2 z} \quad (19)$$

Eq. (6) is rewritten as

$$DrR'' + (1 - Dar)R' - K_2 rR = 0 \quad (20)$$

Because $P = Dr$, $Q = (1 - Dar)$, $S = -K_2 r$, and let $P = Dr = 0$ $D \neq 0$, we know $r = 0$. Thus, we have

$$\frac{Q}{P} = \frac{1 - Dar}{Dr} \ \text{ and } \ \frac{S}{P} = \frac{-K_2 r}{Dr} = -\frac{K_2}{D} \quad (21)$$

To determine whether r = 0 is a regular or irregular singular point, look at

$$(r - 0)\frac{Q}{P} = r\frac{1 - Dar}{Dr} = \frac{1}{D} - ar \quad (22)$$

and

$$(r - 0)^2 \frac{S}{P} = r^2 \frac{-K_2 r}{Dr} = -\frac{K_2 r^2}{D} \quad (23)$$

Both (r-0)[Q(r)/P(r)] and $(r\text{-}0)^2$[S(r)/P(r)] are analytical

at r = 0. Thus, $r = 0$ is a regular singular point of Eq. (20). If r = 0 is a regular singular point, we attempt to obtain a Frobenius series solution, which is a series of the form

$$R = \sum_{n=0}^{\infty} b_n r^{n+q}, \quad b_0 \neq 0 \tag{24}$$

where n is real constant and $q = b_1, b_2, \ldots\ldots$ are known multiples of b_0, which remains arbitrary. b_n is coefficient. The physical unit of r is m.

Thus, the first and second derivatives of a Frobenius series are, respectively,

$$R' = \sum_{n=0}^{\infty} (n+q) b_n r^{n+q-1} \tag{25}$$

and

$$R'' = \sum_{n=0}^{\infty} (n+q)(n+q-1) b_n r^{n+q-2} \tag{26}$$

Substitute Eqs. (25) and (26) into Eq. (20) to get

$$\begin{aligned} &D\sum_{n=0}^{\infty} (n+q)(n+q-1) b_n r^{n+q-1} + \sum_{n=0}^{\infty} (n+q) b_n r^{n+q-1} \\ &-Da\sum_{n=0}^{\infty} (n+q) b_n r^{n+q} - K_2 \sum_{n=0}^{\infty} b_n r^{n+q+1} = 0 \end{aligned} \tag{27}$$

Shift indices in the third and fourth summations to Eq. (27) to obtain

$$\begin{aligned} &D\sum_{n=0}^{\infty} (n+q)(n+q-1) b_n r^{n+q-1} + \sum_{n=0}^{\infty} (n+q) b_n r^{n+q-1} \\ &-Da\sum_{n=1}^{\infty} (n+q-1) b_{n-1} r^{n+q-1} - K_2 \sum_{n=2}^{\infty} b_{n-2} r^{n+q-1} = 0 \end{aligned} \tag{28}$$

Combine these series under one summation, from n = 2, and write the terms corresponding to n = 0 and n = 1 separately, to get

$$\begin{aligned} &Dq(q-1) b_0 r^{q-1} + Dq(q+1) b_1 r^q + q b_0 r^{q-1} + (q+1) b_1 r^q \\ &-Daq b_0 r^q + \sum_{n=2}^{\infty} \left[D(n+q)(n+q-1) b_n + (n+q) b_n \right. \\ &\left. -Da(n+q-1) b_{n-1} - K_2 b_{n-2} \right] r^{n+q-1} = 0 \end{aligned} \tag{29}$$

Let the coefficient of each power of r be zero to get

$$Dq(q-1) b_0 + q b_0 = 0 \tag{30}$$

$$Dq(q+1) b_1 + (q+1) b_1 - Daq b_0 = 0 \tag{31}$$

$$\begin{aligned} &D(n+q)(n+q-1) b_n + (n+q) b_n - Da(n+q-1) b_{n-1} \\ &-K_2 b_{n-2} = 0, (n = 2, 3, 4, \cdots) \end{aligned} \tag{32}$$

Assume that $b_0 \neq 0$. Then Eq. (30) implies that $q = 0$ (It is not suitable.) or $q = 1 - \frac{1}{D}$.

From Eq. (31),we obtain

$$b_1 = \frac{a(D-1)}{2D-1} b_0 \tag{33}$$

From Eq. (30) and $q = 1 - \frac{1}{D}$, we have

$$\begin{aligned} &\left[D\left(n+1-\frac{1}{D} \right)\left(n-\frac{1}{D} \right) + \left(n+1-\frac{1}{D} \right) \right] b_n \\ &-Da\left(n-\frac{1}{D} \right) b_{n-1} - K_2 b_{n-2} = 0 \end{aligned} \tag{34}$$

Eq. (34) can be written as a recurrence relation

$$b_n = \frac{K_2 b_{n-2} + Da\left(n-\frac{1}{D} \right) b_{n-1}}{D\left(n+1-\frac{1}{D} \right)\left(n-\frac{1}{D} \right) + \left(n+1-\frac{1}{D} \right)}, \quad n = 2, 3, 4 \cdots \tag{35}$$

Therefore, substituting n = 2 into Eq. (35) and using Eq. (33), we have

$$\begin{aligned} b_2 &= \frac{K_2 b_0 + Da\left(2-\frac{1}{D} \right) b_1}{6D-2} = \frac{1}{2(3D-1)}\left(K_2 + \frac{(2Da-a)(aD-a)}{2D-1} \right) b_0 \\ &= \frac{a^2(D-1) + K_2}{2(3D-1)} b_0 \end{aligned} \tag{36}$$

Inserting n = 3 into Eq. (35) and using Eqs. (33) and (36), we obtain

$$b_3 = \frac{K_2 b_1 + Da\left(3-\frac{1}{D} \right) b_2}{12D-3} = \frac{a(4D-3)K_2 + a^3(D-1)(2D-1)}{6(2D-1)(4D-1)} b_0 \tag{37}$$

We may continuously derive b_4, b_5, etc. thus, Eq. (24) can be expressed as

$$R(r) = \sum_{n=0}^{\infty} b_n r^{n+1}, \quad n = 0,1,2,3,\cdots$$
$$= b_0 r + \sum_{n=1}^{\infty} b_n r^{n+1}, \quad n = 1,2,3,\cdots \tag{38}$$

Substituting Eqs. (11), (19) and (38) into Eq. (2), we obtain the analytical solution of Eq. (1) as follows

$$C(r,z,t) = C_0 T(0) e^{m_2 z} e^{-K_1 t} \sum_{n=0}^{\infty} b_n r^{n+1} \tag{39}$$

where T(0) is the time function at t = 0, $m_2 = \frac{a - \sqrt{a^2 - 4\frac{(K_1 + K_2)}{D}}}{2}$, K_1 and K_2 are the separation constants, b_n is the coefficients, and n is the exponent of r.

In the case of Eq. (39), if t approaches infinite (∞) or r accesses to zero, then $C(r, z, t) = 0$. If z is larger, then $C(r, z, t)$ is increased. If both z and t approach infinite, then $C(r, z, t) = 0$. Based on the limiting cases of $C(r, z, t)$, it is obvious that Eq. (39) is correct. Further, Eq. (39) is an approximate solution of Eq. (1) due to Eq. (24) which is a Frobenius series solution.

3. Results and Discussion

The analytical solution of the Nernst-Planck equation in unidirectional space and time coordinates is described by [21]. As to the analytical solution of the Nernst-Planck equation in two-dimensional space and time coordinates, some parameters are assumed. The value of T(0) in Eq. (39) represents the time for chloride-ion to pass through concrete specimen, i.e. non-steady-state time. According to Castellote et al. [23], T(0) is called time-lag. Based on the results of available experimental data [13], $T(0)_{CC}$ = 35 hr, $T(0)_{CS}$ = 70 hr, $T(0)_{CF}$ = 90 hr and $T(0)_{CM}$ = 43 hr. The subscripts of CC, CS, CF, and CM concrete were defined are the mix proportion of concrete were free of any mineral blend, in mix 40% of cement was replaced by slag (40%), fly ash (40%), and fly as (12%) and slag (28%), respectively. The separation constants K_1 and K_2 in $m_2 = \frac{a - \sqrt{a^2 - 4\frac{(K_1 + K_2)}{D}}}{2}$ are assumed that equal (The values of K_1 and K_2 are listed in Tables 3-10, 3-11, 3-12 and 3-13 of Reference [21]). It needs to point out that the values of parameters and materials used in the numerical calculation predicted by Eq. (39) are precisely listed in References [21] and [13], respectively. The value of r is 5 cm which is the radius of concrete specimen while the value of b_0 is 10 cm which is the diameter of concrete specimen. We put z = 3 cm and n = 2 into Eq. (39). It is worthy to point out that the method of try and error should be used to do cuve-fitting compared with the experimental results for justifying the properness of selecting an appropriate value of n.

In the case of single set, based on t = 150 hr, Figure 1 shows that $C_{CC(measured)}$ = 0.113 mole L^{-1} > $C_{CC(2\text{-}D\ cylindrical\ coordinates)}$ = 0.1074 mole L^{-1} > $C_{CC(1\text{-}D\ space\ coordinates)}$ = 0.0713 mole L^{-1}. Figure 2 indicates that $C_{CS(measured)}$ = 0.0355 mole L^{-1} > $C_{CS(2\text{-}D\ cylindrical\ coordinates)}$ = 0.0352 mole L^{-1} > $C_{CS(1\text{-}D\ space\ coordinates)}$ = 0.0284 mole L^{-1}. Figure 3 denotes that $C_{CF(2\text{-}D\ cylindrical\ coordinates)}$ = 0.0163 mole L^{-1} > $C_{CF(measured)}$ = 0.0147 mole L^{-1} > $C_{CS(1\text{-}D\ space\ coordinates)}$ = 0.0118 mole L^{-1}. Figure 4 reveals that $C_{CM(2\text{-}D\ cylindrical\ coordinates)}$ = 0.0438 mole L^{-1} > $C_{CM(measured)}$ = 0.0418 mole L^{-1} > $C_{CM(1\text{-}D\ space\ coordinates)}$ = 0.0349 mole L^{-1}. Through the relationship between chloride-ion concentration and time,

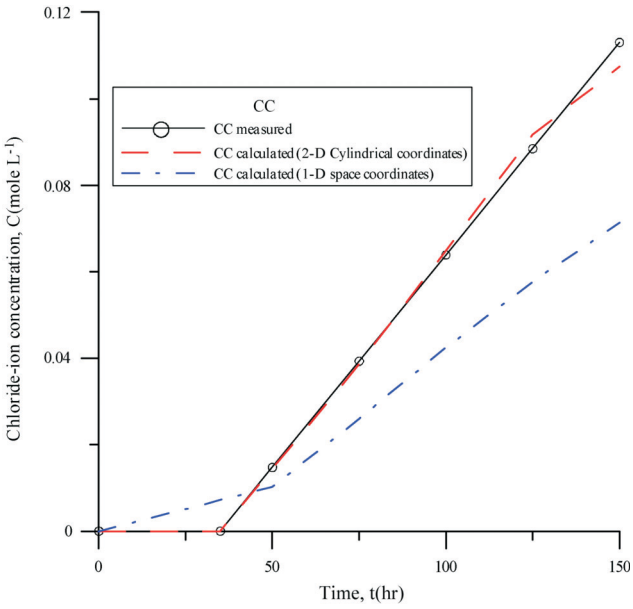

Figure 1. Variations of chloride-ion concentration at steady-state in anode cell for CC concrete.

the analytical results obtained from the Nernst-Planck equation in two-dimensional space and time coordinates more approach the measured results than those of results calculated by the Nernst-Planck equation in unidirectional space and time coordinates [13]. It is evident that the analytical solution of the Nernst-Planck equation in two-dimensional cylindrical space and time coordinates may be in good agreement with the available experimental result obtained from testing cylindrical concrete specimen [13].

On the basis of assumption that $K_1 = K_2$, we know that the separation constants K_1 and K_2 are inversely proportional to time t and $K_{1CC} \rangle K_{1CM} \rangle K_{1CS} \rangle K_{1CF}$. Figures 5–8 show the relationships between time and chloride-

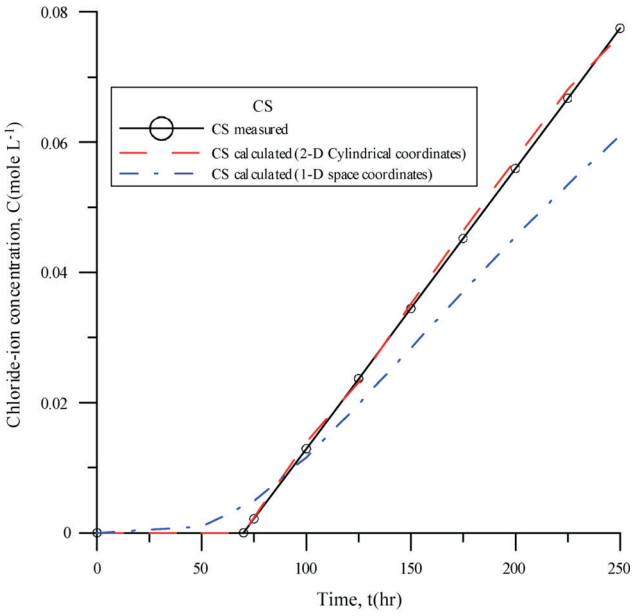

Figure 2. Variations of chloride-ion concentration at steady-state in anode cell for CS concrete.

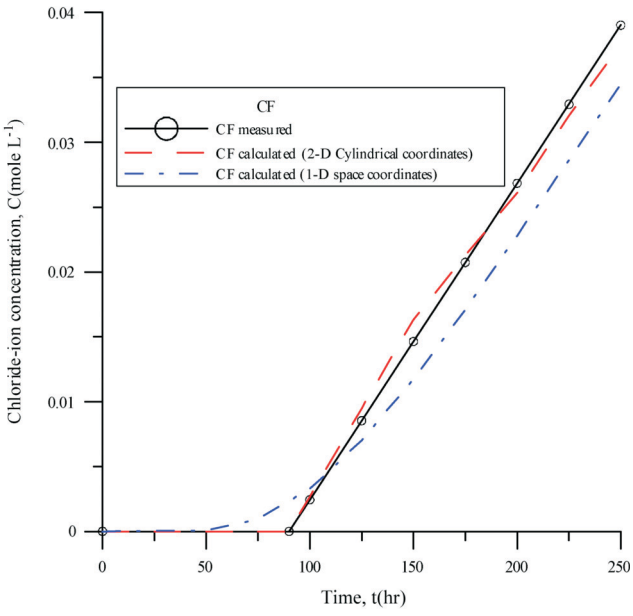

Figure 3. Variations of chloride-ion concentration at steady-state in anode cell for CF concrete.

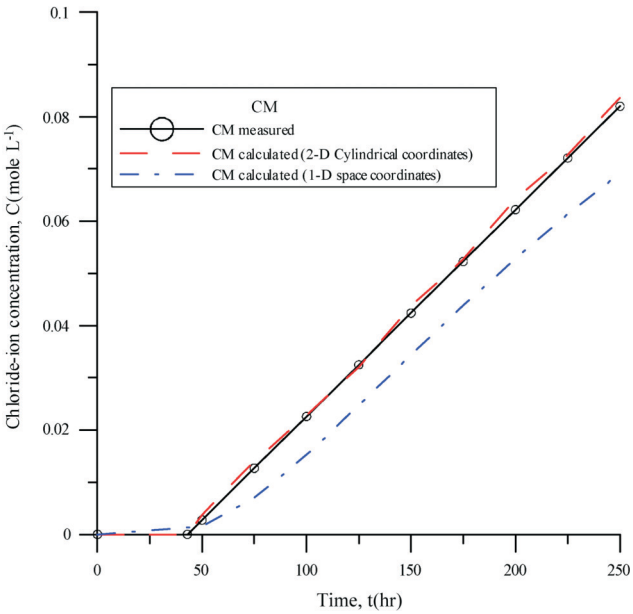

Figure 4. Variations of chloride-ion concentration at steady-state in anode cell for CM concrete.

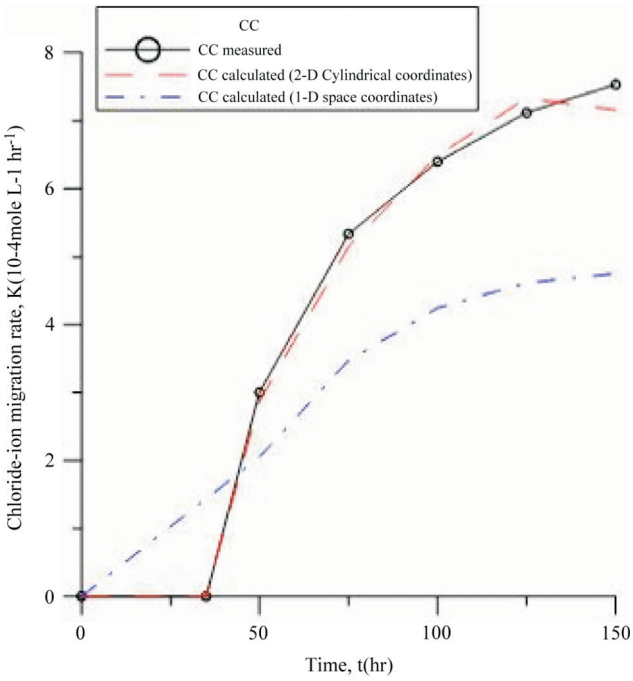

Figure 5. Correlation between time and chloride-ion migration rate for CC concrete.

ion migration rate, K, for CC, CS, CF, and CM concretes, respectively. Using t = 150 hr for comparison, we found that $K_{CC(2\text{-D cylindrical coordinates})} = 7.1583 \times 10^{-4}$ mole $L^{-1}hr^{-1}$ > $K_{CM(2\text{-D cylindrical coordinates})} = 2.5680 \times 10^{-4}$ mole L^{-1} hr^{-1} > $K_{CS(2\text{-D cylindrical coordinates})} = 2.3439 \times 10^{-4}$ mole L^{-1} hr^{-1} > $K_{CF(2\text{-D cylindrical coordinates})} = 1.0856 \times 10^{-4}$ mole L^{-1} hr^{-1}. These analytical and available experimental results show that the CF set is the best to resist the chloride-ion penetration into concrete. The second and third ranking are the CS and CM sets, respectively. The worst one is the CC set. The results of this investigation show that the mineral admixture in ordinary Portland cement (OPC) concrete considerably reduces the diffusion/migration rate for chloride-ion [24].

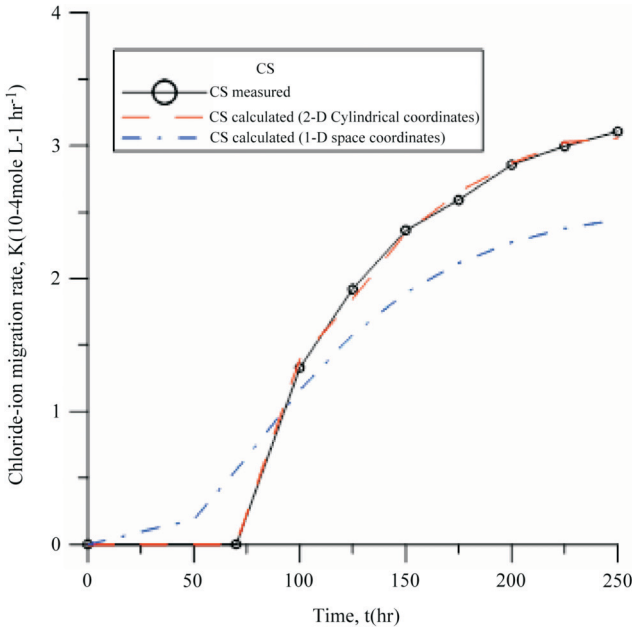

Figure 6. Correlation between time and chloride-ion migration rate for CS concrete.

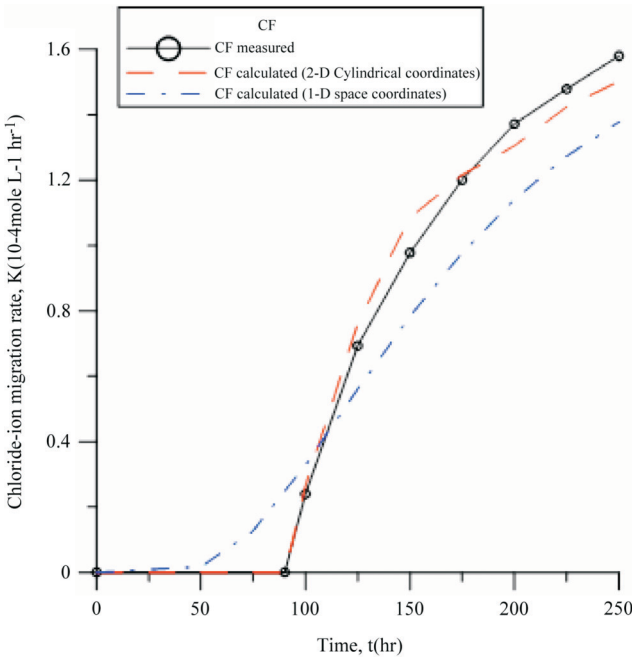

Figure 7. Correlation between time and chloride-ion migration rate for CF concrete.

According to the theoretical background in Ref. [21] and using the experimental data obtained from the available ACMT [13], the effective chloride-ion migration coefficients, D_{eff}, versus initial current, C_u, net charge passed at nonsteady-state, Q_n, and net charge passed at steady-state, Q_s are shown in Figures 9–11, respectively. The corresponding statistical linear regression formulas and correlation coefficients relating initial current, C_u, net charge passed at nonsteady-state, Q_n, and net charge passed at steady-state, Q_s, to effective chloride-ion migration coefficient, D_{eff}, are listed in Tables 1–3, respectively. From Tables 1–3, we found that the values of correlation coefficients of Q_s are better than those of C_u and Q_n. Accordingly, we may use the effective migration coefficient of chloride-ion, D_{eff}, or the net charge passed at steady-state, Q_s, as a durability index for concrete or RC structures.

4. Conclusions

Based on the theoretical analysis and the available

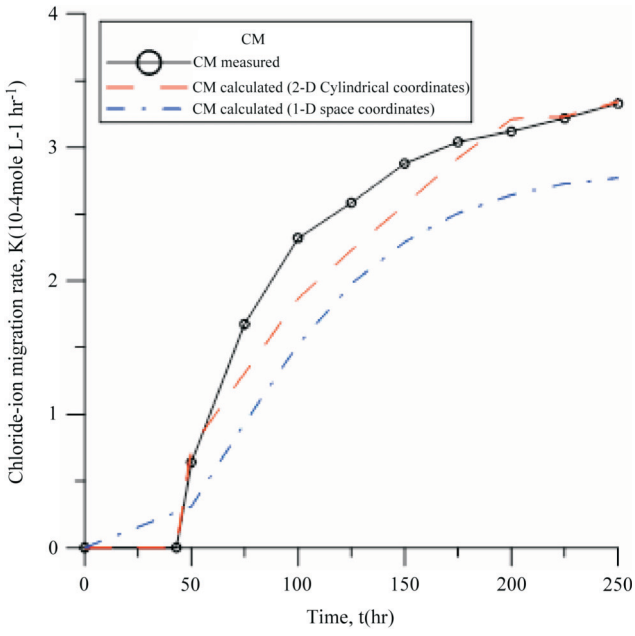

Figure 8. Correlation between time and chloride-ion migration rate for CM concrete.

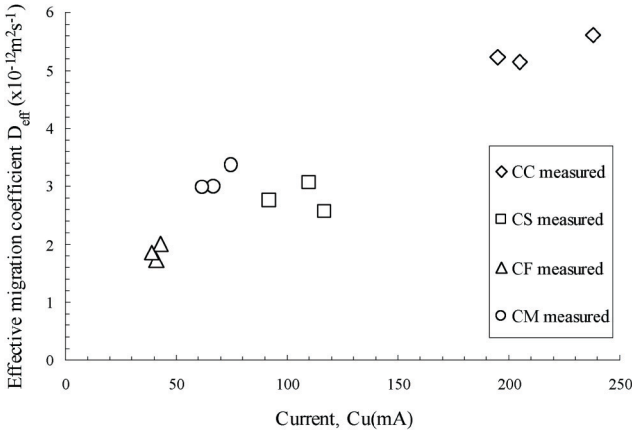

Figure 9. Effective migration coefficient vs. initial current.

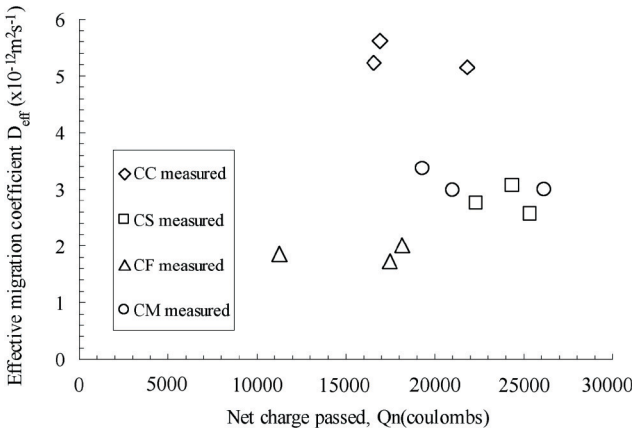

Figure 10. Effective migration coefficient vs. net charge passed at nonsteady-state.

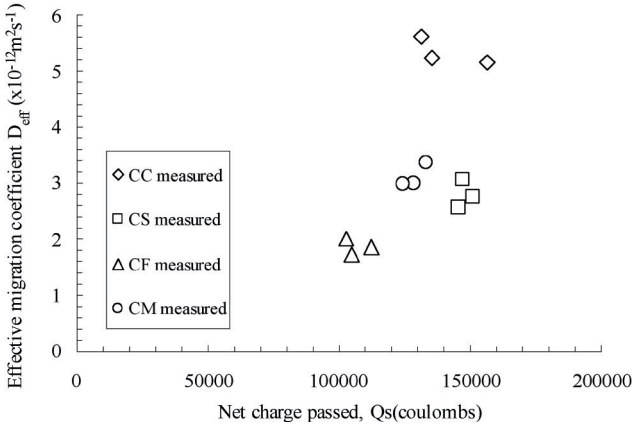

Figure 11. Effective migration coefficient vs. net charge passed at steady-state.

experimental results, the following conclusions can be drawn.

(1) Comparing the measured results between chloride-ion concentration and time obtained from the literature [13] with the calculated results, the Nernst-Planck equation in two-dimensional cylindrical space and time coordinates may be better than the Nernst-Planck equation in unidirectional space and time coordinates.

(2) Using the proposed method, the time-lag, T(0), may be specifically presented the nonsteady-state time during experimental process. However, the value of T(0) cannot be expressed by the Nernst-Planck equation in unidirectional space and time coordinates.

(3) The mineral admixtures such as fly ash and slag in fact can resist the chloride-ion penetration into concrete or RC structures. Particularly, the replacement of fly ash is better than that of slag.

(4) We may adopt the effective migration coefficient of

Table 1. Statistical linear regression formulas and correlation coefficients related initial current to effective chloride-ion migration coefficient

	D_{eff} (10^{-12} $m^2 \times s^{-1}$)	Correlation coefficients R^2
CC measured	$0.0101C_u - 3.1795$	0.8572
CS measured	$-0.0027C_u + 3.0745$	0.0187
CF measured	$0.0375C_u + 0.3292$	0.2865
CM measured	$0.0308C_u + 1.0147$	0.8705

Table 2. Statistical linear regression formulas and correlation coefficients related net charge passed at nonsteady-state to effective chloride-ion migration coefficient

	D_{eff} (10^{-12} $m^2 \times s^{-1}$)	Correlation coefficient R^2
CC measured	-0.00005Qn + 6.2326	0.3433
CS measured	-0.00003Qn + 3.5325	0.0358
CF measured	0.000005Qn + 1.7900	0.0177
CM measured	0.00004Qn + 4.0198	0.4551

Table 3. Statistical linear regression formulas and correlation coefficients related net charge passed at steady-state to effective chloride-ion migration coefficient

	D_{eff} (10^{-12} $m^2 \times s^{-1}$)	Correlation coefficients R^2
CC measured	0.0015Qs – 2.0594	0.9757
CS measured	0.0017Qs – 2.0594	0.7537
CF measured	0.0016Qs – 0.9823	0.8963
CM measured	0.0013Qs – 1.6910	0.9563

chloride-ion, D_{eff}, or the net charge passed at steady-state, Q_s, as a durability index for concrete or RC structures. To justify the correctness and practicability of this conclusion, much more theoretical and experimental research is needed.

If we want to convince to assess the suitability and superiority of Eq. (39) on the predicted transport phenomenon of chloride-ion in concrete, we need much more experimental data compared with the calculated results obtained from Eq. (39).

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